Supporting Information

High performance thermoelectrics from low-cost and abundant CuS/CuI composites

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Additional information

Employed basis sets listed in CRYSTAL17 input format, taken from https://www.crystal.unito.it/basis-sets.php (accessed 14/07/2022).

Copper	**Sulphur**
86-4111(41D)G_doll_2000	86-311G*_lichanot_1993
29 8	16 6
0 0 8 2.0 1.0	0 0 8 2.0 1.0
398000.0 0.000227	109211.0 0.0002520
56670.0 0.001929	16235.206 0.0019934
12010.0 0.01114	3573.0286 0.0111177
3139.0 0.05013	943.23811 0.0498945
947.2 0.17031	287.26179 0.1661455
327.68 0.3693	99.914226 0.3627018
128.39 0.4030	38.602137 0.4108787
53.63 0.1437	15.531224 0.1457875
0 1 6 8.0 1.0	0 1 6 8.0 1.0
1022.0 -0.00487 0.00850	281.22171 -0.0057780
238.9 -0.0674 0.06063	0.0081427
80.00 -0.1242 0.2118	67.106575 -0.0665855
31.86 0.2466 0.3907	0.0565570
13.33 0.672 0.3964	21.794135 -0.1203552
4.442 0.289 0.261	0.2039582
0 1 4 8.0 1.0	8.2097646 0.2741310
54.7 0.0119 -0.0288	0.3973328
23.26 -0.146 -0.0741	3.4178289 0.6463829
9.92 -0.750 0.182	0.3946313
4.013 1.031 1.280	1.5452225 0.2925792
0 1 1 1.0 1.0	0.1544345
1.582 1.0 1.0	0 1 3 6.0 1.0
0 1 1 0.0 1.0	4.3752432 -0.1750000 -

```
 0.596 1.0 1.0
0 1 1 0.0 1.0
 0.150 1.0 1.0
0 3 4 10.0 1.0
 48.54 0.031
 13.55 0.162
 4.52 0.378
 1.47 0.459
0 3 1 0.0 1.0
 0.392 1.0
```

```
0.0613439
 1.8096201 -0.5938952
0.1272251
 0.6833985 0.8298996
1.2215893
0 1 1 0.0 1.0
 0.2413 1.0 1.0
0 1 1 0.0 1.0
 0.106 1.0 1.0
0 3 1 0. 1.
 0.383 1.0
```

Iodide
LC_doll_1998

```
253 5
INPUT
7. 0 2 2 2 1 0
 3.511200    83.113863  0
 1.755600     5.201876  0
 2.968800    82.811109  0
 1.484400     3.379682  0
 1.906600    10.304277  0
 0.953300     7.588032  0
 2.307500   -21.477936  0
0 1 1 2.0 1.0    merged s and p
 2.130007   1.0   1.0
0 1 1 0.0 1.0    merged s and p
 0.3245     1.0   1.0
0 1 1 0.0 1.0    merged s and p
 0.1115     1.0   1.0
0  2  1  5.  1
 2.432887  1.
0  0  1  0.  1
 1.770481  1.
```

Iodide
Mike Towler

```
53 11
0 0 9 2.0 1.11463
 3796580.0      0.0000486
 556463.0      0.000394
 120030.0      0.00229
 30877.7      0.011153
 8938.77      0.046196
 2853.41      0.153983
 1013.04      0.3495
 403.42      0.4298
 169.683      0.20006
0 1 7 8.0 1.13163
 10780.9     -0.000264
0.0010711
 2502.22    -0.00608  0.00978
 769.087    -0.0513   0.0575
 274.618    -0.147205 0.2194
 111.365     0.1122   0.4605
 51.8482     0.5808   0.4568
 25.1274     0.471    0.215
0 1 6 8.0 1.15719
 230.902     0.00648 -0.0135
 91.8899    -0.0261  -0.0666
 38.3274    -0.3172   0.0089
 17.3859    -0.0365   0.8289
 8.1559     0.90233   1.3049
 3.88355    0.4588    0.443
0 3 6 10.0 1.0
 407.397     0.013328
 121.623     0.0935
 45.6127     0.3035
 18.9877     0.470024
 8.4634     0.3103
 3.6279     0.054
0 1 3 8.0 1.0
 9.05308 -2.3837  -0.0815
 4.9116  -0.5412   0.3104
```

```
     2.3444   5.3892   0.719
0 1 1 7.0 1.0
     1.179    1.0      1.0
0 3 3 10.0 1.0
     6.7601   0.2229
     2.7113   0.5615
     1.1174   0.4214
0 3 1 0.0 1.0
     0.4254   1.0
0 1 1 0.0 1.0
     0.585    1.0      1.0
0 1 1 0.0 1.0
     0.2194   1.0      1.0
0 1 1 0.0 1.0
     0.0686   1.0      1.0
```

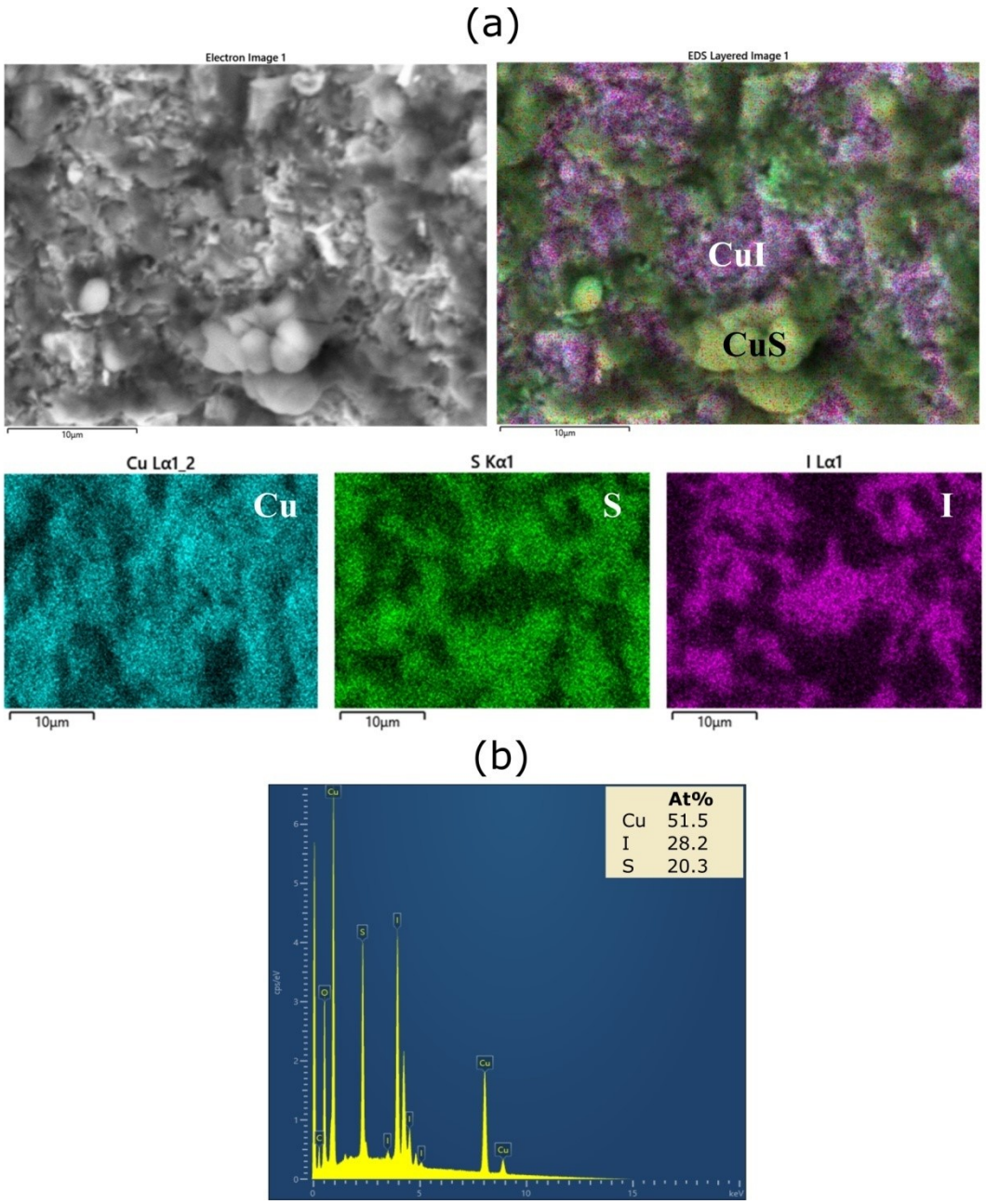

Figure S1. (a) SEM-EDX elemental mapping of a broken solid pellet of CuS:CuI (wt%)-1:4 sample. Images show coloured elemental mapping, (b) shows the composition of the same sample.

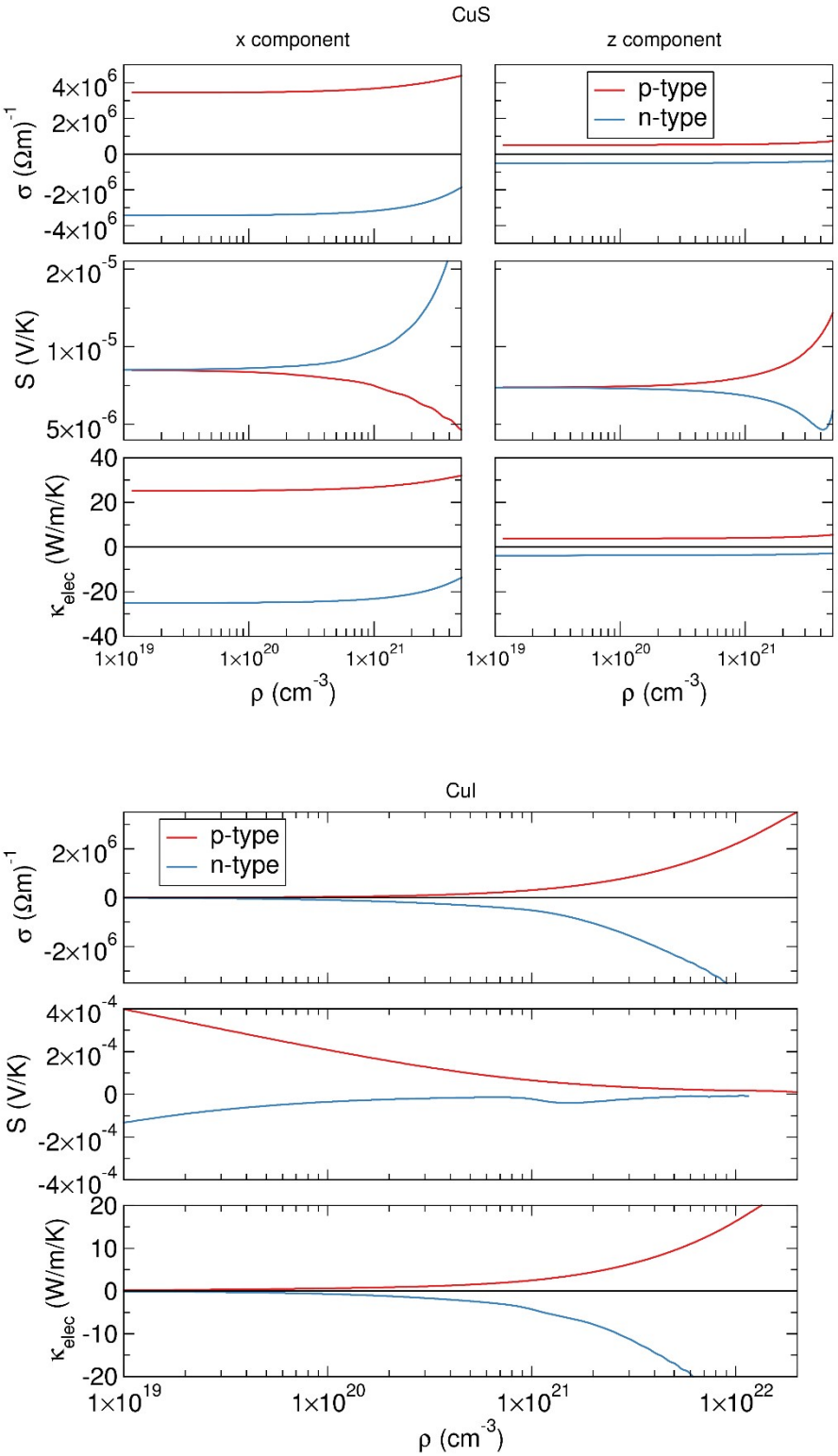

Figure S2. Calculated electrical conductivity (σ), Seebeck coefficient (S), and electronic part of the thermal conductivity (κ) as function of charge carrier concentration of CuS (top) and CuI (bottom) using the HSE functional.

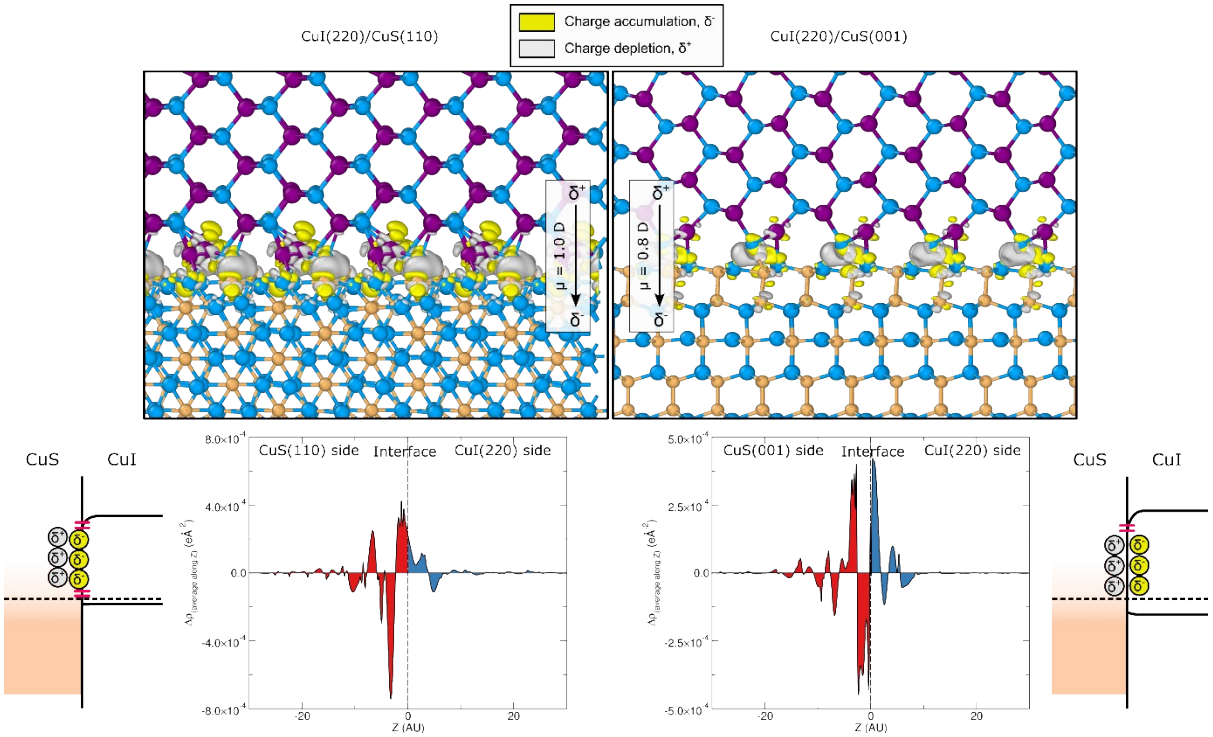

Figure S3. Calculated charge density difference (isosurface reported for a value of 0.002 eÅ^{-3}) together with the band alignment derived from the LPDOS analysis outlined in the main text, supplemented with the band bending mechanism elaborated through the planar averaged charge density.